Application of response surface methodology (RSM) for optimization of strontium sorption by synthetic PPy/perlite nanocomposite

Hossein Esfandian^a, Ahmad Akrami^{b,*}, Fatemeh Bagheban Shahri^b

^aFaculty of Chemical Engineering, Gas and Petroleum, Semnan University, Semnan, Iran ^bDepartment of Chemistry, Tehran Medical Sciences Branch, Islamic Azad University, Tehran, Iran email: aahmad.akramii@gmail.com (A. Akrami)

Received 30 May 2017; Accepted 22 September 2017

ABSTRACT

The aim of this study is to examine the possibility of strontium (Sr) removal from aqueous solution in batch process by synthesized functional polypyrrol/perlite nanocomposite (PPy/Perlite). Themorphology and functional groups of the nanocomposites were characterized by scanning electron microscope (SEM) and Fourier-transform infrared spectroscopy (FTIR). Response surface methodology (RSM) was used to optimize the strontium removal efficiency (%) with three experimental factors using central composite design (CCD). The optimum pH, contact time and amount of adsorbent were 5.12, 28.41 min and 0.23 g, respectively. The maximum removal efficiency of strontium in batch administrations was 97.10%.

Keywords: Strontium; Nanocomposite; Adsorption; RSM; Central composite design; Optimization

1. Introduction

Radio strontium (Sr) interventions in the environment are found as ⁸⁹Sr and ⁹⁰Sr with half-lives of 51 d and 29 y, respectively. It is synthesized as a fission product from nuclear waste repositories and nuclear power plants [1]. Sr-90 is a major radionuclide as part of the nuclear fuel cycle that is a soft β -emitter of 0.5460 MeV energy. Most of the ⁹⁰Sr present in the environment can deposit alone with rain or other condensed moisture. The elemental forms of strontium goes through decomposition while β -radiation and forms ⁹⁰Y with a half-life of 64 h which is a very strong β -emitter. Strontium-90 is comparatively mobile and can move down with water to underlying layers of soil, and the rest remains into groundwater around the world due to the fallout from past atmospheric nuclear weapons tests [2,3]. Because of its chemical similarity with calcium, it can easily adsorb via the gastrointestinal tract. In addition, it accumulates in the body, becomes part of the bone marrow tissue and damages the blood-producing cells [4]. Therefore, ⁹⁰Sr, among all other fission products, is considered as one

*Corresponding author.

of the environmentally hazardous constituents [5]. Thus sorption of Sr from radionuclides should be described as a essential operation before the final disposal of high level liquid waste [2,6].

In recent decades, lots of literature focused on the physical and chemical properties of conducting polymers [7]. Due to their environmental stability, high electrical conductivity and ease of preparation [8], polyaniline and polypyrrole are considerably used in various industries including optics, biosensors [9] and as adsorbent [10,11]. The ion exchange capacity of conducting polymers strongly depends on the polymerization conditions, type and size of the dopants incorporated during the polymerization process [12]. Synthesis of polypyrrole with small dopants i.e. Cl⁻, ClO₄⁻ etc, have considerable anion-exchange behavior due to high mobility of these ions in the polymer matrix [13]. Among different strategies for treatment of radioactive wastes, adsorption process is described as one of the most efficient physical methods for the elimination of pollutants from the environment due to ease of application and cost-effectiveness [2,3,14].

Sorption of metal ions by several functionalized polymers based on amines derivatives such as polyacryloni-

1944-3994 / 1944-3986 © 2017 Desalination Publications. All rights reserved.

trile fibers [15], ethylenediamine [16], polyacrylamides [17], poly-4-vinylpyridine [18] PAN/Zeolite Composite [3], aniline formaldehyde condensate [19] etc, were further reported.

Erdal Kaçan [20] reported an application of response surface methodology (RSM) using central composite design (CCD) to optimize strontium adsorption parameters by activated carbon. The effects of some parameters such as pH, temperature, initial strontium concentration and activated carbon (AC) dosage were studied onto sorption efficiency for strontium adsorption. The experimental results showed very good adsorption performance and high regeneration efficiency of sorbent. In another study, an attempt was made to study the possibility of using PAN/zeolite composite for strontium sorption from aqueous solutions by batch sorption method [3]. Magnetic Fe₃O₄ particles modified sawdust was also used as sorbent for removal of strontium from aqueous solutions. The effect of various operating variables on the sorption of strontium onto Magnetic Fe₃O₄ particles modified sawdust were also investigated [4].

The aim of the present study was to present a facile method to synthesize PPy/perlite nanocomposite. The crystal structure, morphology and chemical structure were systematically and carefully studied by various technologies. PPy/perlite nanocomposite was utilized as an attractive adsorbent for the elimination of strontium from aqueous solution. Moreover, the strontium sorption from aqueous solution in a batch system was studied by RSM package using the Design-Expert software as useful tools to optimize the variables of strontium sorption onto nanocomposite.

2. Materials and methods

2.1. Reagents and chemicals

The adsorbate, i.e. strontium solutions, was prepared by dissolving a known quantity of analytical-grade strontium chloride (Merck,Germany). The pH of solution was adjusted by HCl (1 M) and NaOH (1 M). The concentration of strontium was determined using anatomic absorption spectrophotometer technique (Model 929 Unicam, United Kingdom (UK)). The zeolite (perlite) was purchased from Afrazand Company, Iran. The source of this perlite was from Meyaneh, Iran. The polypyrrole monomer was purchased from Merck (Germany).

2.2. Synthesis of PPy/perlite nanocomposite

The starting material used in this experiment was raw perlite with a grain size of <75 mm (72.68 wt% SiO₂; 11.74 wt% Al₂O₃). In the first step, five grams of perlite were stirred in 100 mL of 0.06 N H₂SO₄ at 87°C for 2 h. The solid was washed with deionized water several times and dried at 110°C overnight.

In the next step, 5 g FeCl₃ was added to 100 mL of deionized water under stirring. Then, 1 g of perlite and 1 mL of fresh distilled pyrrole monomer were added to the stirred solution. The reaction was performed at room temperature for 5 h. Finally, the product was filtered and washed several times with deionized water. The samples were dried at room temperature.

2.3. Instrumentation

In this study, X-ray diffraction (XRD) patterns (MMA, GBC Scientific Equipment LLC, Hampshire, IL, USA) were used to analyze the crystalline structure of perlite through CuK α radiation with a 2 θ scan range 5–80° at room temperature. The morphology and the size distribution of the obtained products were investigated by a scanning electron microscope SEM (model S3400, Hitachi, Japan). The functional groups of the samples were analyzed by Fourier-transform infrared (FTIR) spectrophotometer (Perkin-Elmer, USA). FTIR spectra were recorded by a vector 22 spectrometer, in the range of 400-4000 cm⁻¹ and by diluting a few milligrams of sample in KBr. The spectra were corrected by subtracting the spectrum of a KBr blank pellet and were presented in the transmittance mode. For measuring magnetic properties, room temperature isothermal magnetization between -100 and 100 kOe was carried out using a vibrating sample magnetometer (VSM, MDK6).X-ray fluorescence (XRF, PHILIPS, PW1480, Nederland) was used to characterize perlite elements.

2.4. Adsorption studies

Sorption of strontium was investigated by batch system to examine the effect of various parameters (pH, contact time and amount of adsorbent) on the removal efficiency. For each experiment, 100 ml (50 ppm) of the strontium solution was prepared from the dilution of 1 g/L stock solutions. All experiments were carried out at room temperature on a magnetic stirrer at 400 rpm. At the end of each experiment, the mixture was filtered and the residual strontium ion concentration was analyzed by an atomic absorption spectrophotometer. Each experiment was carried out in duplicate to obtain the reproducibility rate and the mean value used for each set of values. The experimental error was below 5% and no significant differences were observed in the experiment results. The removal efficiency was obtained as follows:

Removal efficiency(%) =
$$\frac{(C_i - C_e)}{C_i} \times 100$$
 (1)

where C_i and C_e are the initial and the equilibrium concentration of strontium, respectively (mg/L).

Adsorption capacity (q) indicates the amount of ions adsorbed per specific amount of sorbent (mg/g). The sorption capacity was obtained as follows:

$$q = (C_i - C_e) \times \frac{V}{m}$$
⁽²⁾

where *q* is the amount of adsorbed strontium (mg/g); *V* is the volume of solution (L); C_i and C_e are the initial and the equilibrium concentration of strontium, respectively (mg/L); and *m* is the weight of adsorbent (g).

2.5. Experimental design

The strontium removal from aqueous solution in a batch system was obtained by RSM package using the Design-Expert software 7.01 (Stat-Ease Inc., Minneapolis, MN, USA) [31]. RSM was used in this study to remove systematic errors and produce an estimate of the experimental error and minimize the number of experiments [32,33]. The experiments were based on CCD to evaluate the relationship between responses and independent variables and also to optimize the relevant conditions of variables with the aim of predicting the best value of responses. By solving the regression equation at the desired values of the process responses as the optimization criteria, the optimum values of the selected variables were obtained [34]. The experiments were based on CCD to study the combined effects of three independent variables (i.e. pH, initial concentration, and contact time). The independent factor levels were coded as -1 (low), 0 (central point or middle), and 1 (high). The variable range and level are given in Table 1 in coded units resulting from RSM studies [35]. A total of 20 (2k + 2k + 6 = 20) experiments were used in this study to evaluate the effects of the three main independent parameters (pH (A), contact time (B) and amount of adsorbent (C)) on strontium removal efficiency. Table 2 presents the result of the CCD experiments that are obtained through studying the effect of three independent variables together with the predicted mean and observed responses. The results were calculated using the Design Expert software.

2.6. Mathematical modeling

The experimental data were subjected to multiple regression analyses. In addition, the CCD design experimental results were fitted in a non-linear regression method [21]. The experiments revealed that each parameter could only come in three levels and the appropriate model was the quadratic equation:

$$Y = \beta_0 + \sum_{i=1}^{k} \beta_i x_i + \sum_{i=1}^{k} \beta_{ii} x_{ii}^2 + \sum_{i=1}^{k-1} \sum_{j=2}^{k} \beta_{ij} x_i x_j + \varepsilon$$
(3)

where *Y* is the predicted response, $X_{i'}, X_j, \ldots, X_{kj}$ are the parameters, X^2i , X^2j , \ldots , X^2k are the square effects, $X_iX_{i'}$, X_iX_k and X_jX_k are the interaction effects, β_0 is the intercept, β_i ($i = 1, 2, \ldots, k$) is regression coefficients for linear effects, β_{ii} ($i = 1, 2, \ldots, k$) is regression coefficients for squared effects, β_{ij} ($i = 1, 2, \ldots, k$) is regression coefficients for squared effects, β_{ij} ($i = 1, 2, \ldots, k$) is regression coefficients for squared effects, β_{ij} ($i = 1, 2, \ldots, k$; $j = 1, 2, \ldots, k$) is regression coefficients for interaction effects, ε is a random error, and k is the number of studied factors. The model terms were confirmed or declined in terms of the probability (P) value with a 95% confidence level. Using the Design Expert software, the results were completely analyzed via the analysis of variance (ANOVA) [21,22].

3. Results and discussion

3.1. Adsorbent charactisation

Fig. 1 shows the XRD patterns of the raw perlite representing a hump of glass. It indicates that perlite is an amorphous material (volcanic glass). The characteristics of the

Table 1

Experimental ranges and levels of the independent variables

| Type of variable | Name of variables | Range and level | | |
|------------------|----------------------|-----------------|-----|-----|
| | | -1 | 0 | +1 |
| Numerical | pН | 2 | 5 | 8 |
| | Contact time (min) | 10 | 20 | 30 |
| | Amount of dosage (g) | 0.1 | 0.2 | 0.3 |

Table 2

Full factorial central composite design matrix of orthogonal and real values along with observed responses for strontium sorption (%).

| Run | Variable with coded levels | | Removal efficiency (%) | | |
|-----|----------------------------|------------------------------------|----------------------------|--------------|-----------|
| | рН | Amount of Contact time (min) | Amount of dosage (g) | Experimental | Predicted |
| 1 | -1 | -1 | 1 | 43.5 | 40.08 |
| 2 | 1 | 1 | 1 | 47.74 | 49.29 |
| 3 | -1 | 1 | 1 | 58.85 | 60.39 |
| 4 | 1 | 1 | -1 | 23.54 | 22.6 |
| 5 | -1 | -1 | -1 | 30.43 | 28.68 |
| 6 | 0 | 0 | -1 | 67.87 | 74.74 |
| 7 | 0 | 0 | 0 | 94.43 | 92.84 |
| 8 | -1 | 0 | 0 | 50.92 | 54.97 |
| 9 | 0 | 0 | 0 | 94.62 | 92.84 |
| 10 | 1 | -1 | -1 | 21.33 | 17.58 |
| 11 | 0 | 1 | 0 | 95.21 | 93.49 |
| 12 | -1 | 1 | -1 | 34.13 | 43.87 |
| 13 | 1 | 0 | 0 | 43.13 | 39.25 |
| 14 | 1 | -1 | 1 | 26.59 | 28.98 |
| 15 | 0 | 0 | 0 | 94.33 | 92.84 |
| 16 | 0 | 0 | 1 | 95.85 | 93.78 |
| 17 | 0 | -1 | 0 | 74.31 | 80.83 |
| 18 | 0 | 0 | 0 | 94.68 | 92.84 |
| 19 | 0 | 0 | 0 | 94.46 | 92.84 |
| 20 | 0 | 0 | 0 | 94.12 | 92.84 |



Fig. 1. XRD pattern of perlite.

| Characteristics of the raw perlite | | | | | |
|------------------------------------|--------------------|--|--|--|--|
| Element | Composition (%w/w) | | | | |
| SiO ₂ | 72.68 | | | | |
| Al_2O_3 | 11.74 | | | | |
| Fe ₂ O ₃ | 0.85 | | | | |
| CaO | 0.78 | | | | |
| Na ₂ O | 3.52 | | | | |
| K ₂ O | 5.21 | | | | |
| MgO | 0.02 | | | | |
| MoO ₃ | 3.835 | | | | |
| TiO ₂ | 0.069 | | | | |
| MnO | 0.072 | | | | |
| P_2O_5 | 0.008 | | | | |
| S | 0.015 | | | | |
| L.O.I | 0.89 | | | | |

Table 3



Fig. 2. SEM images of PPy/perlite.

sorbent are indicated in Table 3. The SEM of PPy/perlite composites is illustrated in Fig. 2. As can be seen, the spherical nano sized particle has been formed with an average size of 100 nm. The formation of the PPy in the nanocomposites was investigated by FTIR. The spectrum of the composite clearly exhibits characteristic absorption peaks with respect to polypyrrole (Fig. 3). The band at 1558 cm⁻¹ which corresponds to the C–C and C=C stretching of the aromatic ring is characterized. It should also be noted that the C–N stretching (aromatic) at 1400 cm⁻¹ confirms the presence of polypyrrole. The broad band from 1400 to 1000 cm⁻¹ is attributed to C–H or C–N in-plane deformation modes and has a maximum at 1089 cm⁻¹ [23]. The absorptions at 615 cm⁻¹ are related to the C–H outer bending vibrations [7].

3.2. Statistical analysis

The analysis of variance ANOVA based on the RSM approach is a statistical technique that subdivides the total variation in a set of data into component parts associated



Fig. 3. FTIR images of of PPy/perlite.

with specific sources of variation for the purpose of testing hypotheses on the parameters of the model. ANOVA was used to check the significance and fitness of the model. The results related to all responses are shown in Table 4. Based on Eq. (2), the F-values and those of probability >F determined the significance of each coefficient [24]. The large value of F shows that most of the variation in the response can be explained by the regression equation. The associated *p* value is used to estimate whether $F_{\text{Statistics}}$ is large enough to indicate statistical significance. A *p* > F-value, less than 0.05, indicates a high significant regression at 95% confidence level. Values greater than 0.1 show that the model terms are not significant [25]. In this study, the factors of first-order effects and square effects were specified as significant model terms. The regression model for strontium adsorption was found to be highly significant from the Fisher's test having a high F-value (109.9) with very low probability (p<0.0003). The not significant value resulting from the lack of fit (more than 0.05) indicated that the quadratic model was valid for response. The examination of the fit summary output suggested that the quadratic model is statistically significant for the response. Hence it is used for further analysis [21]. The analysis of variance for the three parameters (pH, contact time and amount of adsorbent) indicates that strontium sorption can be well described by a polynomial model with a relatively high correlation coefficient ($R^2 = 0.99$). Table 4 indicates the empirical relationship between responses (Y) and the three variables (factors) in coded values obtained by the application of RSM. The P-values were used as a tool to check the significance of each coefficient, which in turn might represent the pattern of the interactions between the variables. As shown in this table, coefficients (A, B, C, BC, A², B² and C²) were significant with very small P-values (P < 0.05). The other term coefficients were not significant (P > 0.05). For each response, the square of correlation coefficient was computed as the R square (R²) that is a measure of the amount of variation around the mean explained by the model [24,26].

The model coefficient of determination (\mathbb{R}^2) had a very high value (0.99) indicating the statistical significance of this regression and only 0–4% of the total variations are not explained by the model (Table 5). The predicted \mathbb{R}^2 showed the goodness of response. The value of the predicted determination of the coefficient (Pred. $\mathbb{R}^2 = 0.89$) is in reasonable accord with the value of adjusted determination of the coefficient (Adj. $\mathbb{R}^2 = 0.98$). However, a relatively lower value of the coefficient of variance (CV = 6.23%) for response

 Table 4

 An analysis of variance (ANOVA) for the response surface quadratic model

| Source | Sum of | df | Mean | F | p-value | |
|----------------------|----------|----|---------|--------|----------|-------------|
| | Squares | | Square | Value | Prob > F | |
| Model | 15704.96 | 9 | 1745.00 | 109.90 | < 0.0001 | Significant |
| A - pH | 308.32 | 1 | 308.32 | 19.40 | 0.0013 | |
| B - contact time | 400.82 | 1 | 400.82 | 25.24 | 0.0005 | |
| C - Amount of dosage | 906.88 | 1 | 906.88 | 57.11 | < 0.0001 | |
| AB | 2.32 | 1 | 2.32 | 0.15 | 0.7102 | |
| AC | 8.67 | 1 | 8.67 | 0.55 | 0.4768 | |
| BC | 116.97 | 1 | 116.97 | 7.37 | 0.0218 | |
| A ² | 5183.48 | 1 | 5183.48 | 326.45 | < 0.0001 | |
| B ² | 88.74 | 1 | 88.74 | 5.59 | 0.0397 | |
| C^2 | 202.47 | 1 | 202.47 | 12.75 | 0.0051 | |
| Residual | 158.78 | 10 | 15.88 | | | |
| Lack of Fit | 158.58 | 5 | 31.72 | | | |
| Pure Error | 0.21 | 5 | 0.041 | | | |
| Cor. Total | 15863.74 | 19 | | | | |

Table 5

ANOVA results for response parameters

| Standard deviation | 3.98 | R-Squared | 0.99 |
|--------------------|---------|----------------|--------|
| Mean | 64.00 | Adj R-Squared | 0.98 |
| C.V.% | 6.23 | Pred R-Squared | 0.89 |
| PRESS | 1145.59 | Adeq Precision | 26.865 |
| | | | |

suggests a good precision and reliability in the conducted experiments. The final second-order polynomial equation by multiple regressions (Y) and the significant terms (p < 0.05) obtained are as follows:

Removal efficiency
$$(\%)_{s_r} = 92.84 - 5.5A + 6.33B + 9.52C + 3.82BC - 43.42A^2 - 5.68B^2 - 8.58C^2$$
 (4)

Fig. 4 indicates the relationship between actual and predicted values of Y for adsorption of strontium onto nanocomposite. As shown in Fig. 4, developed models are adequate because the residuals for the prediction for most of the responses are minimum, since the residuals tend to be close to the diagonal line.

3.3. Effect of various parameters on strontium sorption

The experimental design models, the central composite design (CCD) model and RSM, were used to evaluate the effects of three important variables on strontium sorption to find the optimum process conditions. Hence, the individual effects of process variables on strontium adsorption were discussed below.

3.3.1 Effect of pH on the response

Solution pH is one of the important operational variables in every wastewater treatment system. Subsequent



Fig. 4. Predicted response vs. actual response.

study of this parameter with the purpose of examining the optimized point of purification or absorbance, yield, and the effect of deviation from this point in work and the analysis of sensitivity of parameter revealed high performances and suggestive results. Fig. 5 shows the combined effects of pH which correspond with contact time and amount of dosage. As can be seen, increasing pH leads to an increase in the strontium sorption; and it later reaches a maximum value at pH 5 which then decreases for nanocomposite. It can be concluded that an increase in the aqueous solution pH leads to an increase in the negative charge density of the adsorbent. Thus, the uptake of the strontium might be ascribed to increasing negative charge on the adsorbent surface (i.e. nanocomposite) [24]. At low pH values, the concentration of H⁺ ions increases, and thus an intense competition occurs between H⁺ ions and strontium ions for sorption on binding sites of PPy/perlite nanocomposite. As a result, H⁺ ions could either occupy the binding sites or protonate functional groups on PPy/perlite, i.e. metal-binding sites. Because of protonation, the amount of positively charged

78



Fig. 5. Diagram of the pH effect on the removal efficiency (%) of strontium.

A: pH



Fig. 6. Contour and 3-D diagram of the effect of pH and contact time (min) on removal efficiency (%).

sites increased. Also, the corresponding electrostatic repulsion restricted the adsorption of positively charged strontium ions. Thus, the sorption capacity of strontium ions reduced at pH 2.0. On the other hand, as the pH values increased, the negative charge density on the surface of PPy/perlite nanocomposite also increased due to deprotonation of the metal binding sites. This in turn enhanced the affinity toward positively charged strontium ions and led to the improved adsorption of metal ions [27,28]. The three-dimensional (3D) response surface plots and counter plots are illustrated in Fig. 6, which represent the expected removal efficiency response and effects of pH and contact time due to the significant interaction between them. Shortly afterwards, the strontium sorption increased at pH = 5 and a 20



Fig. 7. Contour and 3-D diagram of effect of pH and amount of dosage (g) on removal efficiency (%).

min contact time and the maximum removal efficiency, i.e. 95% were achieved. After this point, the sorption capacity decreased. Fig. 7 shows the combined effects of pH corresponding with the amount of dosage, on strontium sorption. As can be seen, when the contact time is constant (20 min), by varying the others variables, the removal efficiency changes in the range of 21 to 95 (%). Maximum removal efficiency of strontium was obtained at pH 5, an amount of dosage of 0.3 g.

3.3.2. Effect of contact time on the response

In every batch treatment system, contact time has a significant effect on the final yield of the system. In the treatment systems related to other processes, the time or waste time in each section may have effects on the system overall capacity and efficacy. In fact, reaction time of every part of the system and the respective sensitivity to deviation have a direct and/or indirect effect on all costs and yields. It should be noted that the contact time in the sorption process is considered as the main parameter for equilibrium. This signifies that the variable was optimized. In addition, the subsequent increase in the point of equilibrium has no effect on the absorption process [21,24,29].

Fig. 8 represents the uptake of strontium by nanocomposite at different contact times. It is obvious that increasing the contact time leads to an improvement in the strontium sorption. At equilibrium, no significant change was observed. Therefore, almost all of the strontium is removed after 20 min. Fig. 9 (3-D and contour diagrams) illustrates maximum removal efficiency of strontium versus contact



Fig. 8. Diagram of the contact time effect on the removal efficiency (%) of strontium.



Fig. 9. Contour and 3-D diagram of the effect of contact time (min) and amount of dosage (g) on removal efficiency (%).

time and amount of dosage. It was observed that at pH 5 (by varying other variables), removal efficiency varied from 21 to 95%. Clearly, an increase in contact time and the amount of adsorbent values lead to an increase in removal efficiency of strontium.

3.3.3. The effect of the amount of adsorbent of the model on the responses

Strontium sorption onto nanocomposite was carried out at three levels of sorbent amount, in the range of 0.1-0.3 g.



Fig. 10. Diagram of the amount of dosage effect on the removal efficiency (%) of strontium.

Fig. 10 shows the effect of quantity of adsorbent on the removal efficiency of strontium. As can be seen, increasing the amount of sorbent leads to a linear increase in the removal yield of strontium in the model. It can be described in this way that by increasing the adsorbent dosage, more adsorption sites can be provided for process; therefore it is expected that the amount of adsorbed strontium ions increase with increasing the sorbent dosage.

3.4. Process optimization

The purpose of the study is optimization of some variables to achieve maximum strontium sorption. The optimized condition is preset at high and low levels or in the ranges of the process variables. Table 6 shows the optimization criteria used to obtain the optimal region for the response. To carry out the optimization phase, the desirability function procedure was used. The desirability function procedure is one of the widely used multi-response optimization methods in practice that locates one or more points that maximize this function. The desirability lies between 0 and 1 and it is indicative of the proximity of a response to its ideal value [24,30]. Table 7 shows the results of the optimum conditions. In Table 7, the best optimum condition of process variables is number 1 which has the highest value of desirability. The results are closely related to the data obtained from optimization analysis using desirability functions, indicating that the CCD design in corporation with desirability functions could be effectively used to optimize the adsorption parameters for the removal of strontium.

Additional experiments were performed at 5 optimum conditions to verify the accuracy. The optimal conditions were produced by the design expert software. The comparison of the experimental values and the software-predicted ones are indicated in Table 8. The results suggest there is a mean error of 2.12%.

3.5. Comparison of the maximum sorption capacity of various sorbents

To have a better understanding of the adsorption capacity of PPy/perlite nanocomposite, the values of the maximum

Table 6 Parameters of the response optimization

| Criteria | Target | Lower limit | Upper limit |
|----------------------|-------------|----------------|----------------|
| pН | Is in range | 4 | 6 |
| Contact time (min) | Is in range | 20 | 30 |
| Amount of dosage (g) | Is in range | 0.2 | 0.3 |
| Strontium removal, % | Maximize | 55 | 307 |

Table 7

Optimum diazinon removal condition by modified zeolite

| No. | pН | Contact time (min) | Amount of dosage (g) | Adsorption capacity (mg/g) | Desirability |
|-----|------|--------------------------|----------------------------|----------------------------------|--------------|
| 1 | 5.12 | 28.41 | 0.23 | 97.10 | 1 |
| 2 | 4.64 | 23.20 | 0.22 | 96.36 | 1 |
| 3 | 4.56 | 23.65 | 0.27 | 97.78 | 1 |
| 4 | 4.13 | 28.02 | 0.28 | 96.9 | 1 |
| 5 | 5.43 | 25.41 | 0.29 | 96.45 | 1 |
| 6 | 4.47 | 22.33 | 0.24 | 96.30 | 1 |
| 7 | 4.95 | 25.12 | 0.22 | 96.30 | 1 |
| 8 | 5.42 | 27.29 | 0.27 | 97.18 | 1 |
| 9 | 4.77 | 24.22 | 0.22 | 96.33 | 1 |
| 10 | 5.50 | 28.34 | 0.26 | 96.40 | 1 |

Table 8 Verification experiments at optimum condition

| Run | Strontium removal (%) | | Error (%) |
|------------|-----------------------|-----------------|-----------|
| | Experimental | Predicted (DOE) | - |
| 1 | 95.18 | 97.10 | 2.02 |
| 2 | 94.13 | 96.36 | 2.37 |
| 3 | 94.43 | 97.78 | 3.55 |
| 4 | 95.12 | 96.9 | 0.81 |
| 5 | 95.34 | 96.45 | 1.87 |
| Mean Error | _ | _ | 2.12 |

sorption obtained for strontium ion uptake with various types of sorbents are shown in Table 9. The experimental data collected from the present investigations correspond to the reported values. As shown in Table 9, the sorption capacity of PPy/perlite nanocomposite is higher than some sorbent such as PAN/Zeolite, sawdust modified Fe3O4 particles, crosslinked polyzwitterion/anion and aerobic granules. However, its performance index is less than those of the brown marine algae and organo-montmorillonites.

4. Conclusion

In this study, a facile and low cost method has been developed to synthesize a novel material, i.e. PPy/perlite

Table 9 Comparison of the maximum sorption capacity (q_m) of the various sorbents

| Sample | Sorbent material | $q_m (\mathrm{mg/g})$ | Reference |
|--------|--|-----------------------|-----------|
| 1 | PPy/perlite nanocomposite | 16.47 | This work |
| 2 | PAN/Zeolite | 0.011 | [31] |
| 3 | $Fe_{3}O_{4}$ particles modified sawdust | 12.59 | [4] |
| 4 | Cross-linked polyzwitterion/anion | 0.365 | [32] |
| 5 | Aerobic granules | 13.65 | [33] |
| 6 | Brown Marine Algae | 330.38 | [5] |
| 7 | Organo-montmorillonites | 65.65 | [34] |

nanocomposite. The formation of polypyrrol at the surface of perlite was confirmed using FT-IR measurements. The SEM image showed that the size of crystalline nanoparticles was about 100 nm. Nanocomposite synthesized can show a better performance for the separation of strontium from wastewater. To optimize the operational conditions for achieving the maximum sorption of strontium, the RSM experimental design was used. The employed variables for wastewater treatment process were pH(A), contact time (min) (B), and amount of dosage (g) (C). These variables were studied in laboratory and the optimum value of each parameter was determined to reach the maximum value of responses. At optimum conditions, a removal efficiency of 95% was obtained. The results of the analysis using the CCD design indicated that the obtained approximation models for strontium were satisfactorily fitted with R² of 0.99.

References

- D. Chakraborty, S. Maji, A. Bandyopadhyay, S. Basu, Biosorp-tion of cesium-137 and strontium-90 by mucilaginous seeds of [1] Ocimum basilicum, Bioresource Technol., 98 (2007) 2949–2952. H. Esfandian, H. Fakhraee, A. Azizi, Removal of strontium
- [2] ions by synthetic nano sodalite zeolite from aqueous solution, Int. J. Eng-Transact. B: Applications, 29 (2016) 160.
- Y. Sabriye, E. Sema, Adsorption characterization of strontium [3] on PAN/zeolite composite adsorbent, World. J. Nuclear Sci. Technol., 1 (2011) 6–12
- Z. Cheng, Z. Gao, W. Ma, Q. Sun, B. Wang, X. Wang, Prepa-[4] ration of magnetic Fe₃O₄ particles modified sawdust as the adsorbent to remove strontium ions, Chem. Eng. J., 209 (2012) 451-457.
- [5] M. Khani, Biosorption of strontium by a nonliving brown
- Marine algae, *Padina sp*, Sep. Sci. Technol., 47 (2012) 1886–1897. A. Zhang, Y. Wei, H. Hoshi, M. Kumagai, Synthesis of a novel silica-based macroporous polymer containing TODGA [6] Chelating agent and its application in the chromatographic separation of Mo (VI) and Zr (IV) from diethylenetriaminepentaacetic acid, Sep. Sci. Technol., 40 (2005) 811–827. P. Chandrasekhar, Conducting polymers, Fundamentals and
- [7] Applications: A Practical Approach., 1999, 83 p.
- [8] P. Chandrasekhar, Conducting polymers, fundamentals and applications: a practical approach, Springer, 1999.
- [9] M. Nishizawa, T. Matsue, I. Uchida, Fabrication of a pH-sensitive microarray electrode and applicability to biosensors, Sensors Actuat. B: Chem., 13 (1993) 53–56.
- B. Saoudi, N. Jammul, M.-L. Abel, M.M. Chehimi, G. Dodin, [10] DNA adsorption onto conducting polypyrrole, Synthetic Met., 87 (1997) 97–103.

- [11] X. Zhang, R. Bai, Surface electric properties of polypyrrole in aqueous solutions, Langmuir, 19 (2003) 10703–10709.
- [12] C. Weidlich, K.-M. Mangold, K. Jüttner, Conducting polymers as ion-exchangers for water purification, Electroch. Acta, 47 (2001) 741–745.
- [13] R. Ansari, Polypyrrole conducting electroactive polymers: synthesis and stability studies, J. Chem., 3 (2006) 186–201.
- [14] S.N. Azizi, N. Asemi, Parameter optimization of the fungicide (Vapam) sorption onto soil modified with clinoptilolite by Taguchi method, J. Environ. Sci. Health Part B., 45 (2010) 766–773.
- [15] S. Deng, R. Bai, J.P. Chen, Aminated polyacrylonitrile fibers for lead and copper removal, Langmuir, 19 (2003) 5058–5064.
- [16] P. Chethan, B. Vishalakshi, Synthesis of ethylenediamine modified chitosan and evaluation for removal of divalent metal ions, Carbohyd. Polym., 97 (2013) 530–536.
- [17] B. Mathew, V.R. Pillai, Polymer-metal complexes of amino functionalized divinylbenzene-crosslinked polyacrylamides, Polymer, 34 (1993) 2650–2658.
- [18] P. Viel, S. Palacin, F. Descours, C. Bureau, F. Le Derf, J. Lyskawa, M. Salle, Electropolymerized poly-4-vinylpyridine for removal of copper from wastewater, Appl. Surf. Sci., 212 (2003) 792–796.
- [19] P.A. Kumar, S. Chakraborty, Fixed-bed column study for hexavalent chromium removal and recovery by short-chain polyaniline synthesized on jute fiber, J. Hazard. Mater., 162 (2009) 1086–1098.
- [20] E. Kaçan, Strontium ion removal from aqueous solution using activated carbon produced from textile sewage sludges: effect of ZnCl₂, Usak Univ. J. Mater. Sci., 3 (2014) 165.
- [21] M.S. Baei, H. Esfandian, A. A. Nesheli, Removal of nitrate from aqueous solutions in batch systems using activated perlite: an application of response surface methodology, Asia-Pacific J. Chem. Eng., 11 (2016) 437–447.
- [22] S. Subramaniam, A. Palanisamy, A. Sivasubramanian, Box-Behnken designed adsorption based elution-unique separation process for commercially important acetyl shikonin from Arnebia nobilis, RSC Adv., 5 (2015) 6265–6270.
- [23] J.W. Choi, K.B. Lee, K.Y. Park, S.Y. Lee, D.J. Kim, Comparison between Ti-and Si-based mesostructures for the removal of phosphorous from aqueous solution, Environ. Prog. Sustain. Energy, 31 (2012) 100–106.
- [24] H. Esfandian, M. Parvini, B. Khoshandam, A. Samadi-Maybodi, Removal of diazinon from aqueous solutions in batch systems using Cu-modified sodalite zeolite: An application of response surface methodology, Int. J. Eng-Transact. B: Applications., 28 (2015) 1552.

- [25] A. Igder, A.A. Rahmani, A. Fazlavi, M.H. Ahmadi, A. Azqhandi, M. Hossein, M.H. Omidi, Box-Behnken design of experiments investigation foradsorption of Cd²⁺ onto carboxymethyl chitosan magnetic nanoparticles, J. Mining. Environ., 3 (2012) 51–59.
- [26] F. Ghorbani, H. Younesi, S.M. Ghasempouri, A.A. Zinatizadeh, M. Amini, A. Daneshi, Application of response surface methodology for optimization of cadmium biosorption in an aqueous solution by Saccharomyces cerevisiae, Chem. Eng. J., 145 (2008) 267–275.
- [27] B. Alizadeh, M. Ghorbani, M.A. Salehi, Application of polyrhodanine modified multi-walled carbon nanotubes for high efficiency removal of Pb(II) from aqueous solution, J. Mol. Liq., 220 (2016) 142–149.
- [28] S.A. Kosa, G. Al-Zhrani, M.A. Salam, Removal of heavy metals from aqueous solutions by multi-walled carbon nanotubes modified with 8-hydroxyquinoline, Chem. Eng. J., 181 (2012) 159–168.
- [29] T.A. Saleh, A. Sarı, M. Tuzen, Optimization of parameters with experimental design for the adsorption of mercury using polyethylenimine modified-activated carbon, J. Environ. Chem. Eng., 5 (2017) 1079–1088.
- [30] S. Raissi, Developing new processes and optimizing performance using response surface methodology, World Academy of Science, Eng. Technol., 49 (2009) 1039–1042.
- [31] S. Yusan, S. Erenturk, Adsorption characterization of strontium on PAN/zeolite composite adsorbent, World. J. Nuclear. Sci. Technol., 1 (2011) 6–12.
- [32] S.A. Ali, S.A. Haladu, A novel cross-linked polyzwitterion/ anion having pH-responsive carboxylate and sulfonate groups for the removal of Sr²⁺ from aqueous solution at low concentrations, Reactive Func. Polym., 73 (2013) 796–804.
- [33] L. Wang, C. Wan, D.-J. Lee, J.-H. Tay, X. Chen, X. Liu, Y. Zhang, Adsorption–desorption of strontium from waters using aerobic granules, J. Taiwan Inst. Chem. Eng., 44 (2013) 454–457.
- [34] P. Wu, Y. Dai, H. Long, N. Zhu, P. Li, J. Wu, Z. Dang, Characterization of organo-montmorillonites and comparison for Sr (II) removal: equilibrium and kinetic studies, Chem. Eng. J., 191 (2012) 288–296.